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Optical second harmonic generation studies of the dc-electric field screening in Si–SiO_2 multiple quantum wells

V. V. Savkin†, A. A. Fedyanin†, F. A. Pudonin‡, A. N. Rubtsov†
and O. A. Aktsipetrov†

† Department of Physics, Moscow State University, 119899, Moscow, Russia
‡ Lebedev Physical Institute of RAS, Leninsky pr. 53,
117924, Moscow, Russia

Abstract. Dc-electric field screening in the Si–SiO_2 multiple quantum wells is studied by optical second harmonic generation. Experimentally observed oscillations in the nonlinear response, inconsistent with semiclassical description of screening, are explained within the Tomas-Fermi approach.

The significant attention has been given recently to the study of the influence of the external electric field on the nonlinear-optical properties of the solids. In particular, the electric field effects in low-dimensional systems such as multiple quantum wells (MQWs) and superlattices are of interest. The optical second-harmonic generation (SHG) is sensitive to the symmetry properties, as a consequence practically there is no SHG signal from the centrosymmetric material. Dc-electric field induced SHG (EFISH) is determined by polarization $P_{2\omega}$ as $P_{2\omega} = \chi^{(3D)}(\omega, \omega, 0)E_\omega E_\omega E_0$, where $\chi^{(3D)}$ is nonlinear susceptibility, E_ω and E_0 are fundamental and dc-electric field. To interpret EFISH results in low-dimensional structures it is important to know the screening of the electric field in the structure.

In this paper, we present the description of screening of dc-electric field in MQWs. The basic method of research of screening electric field in Si–SiO_2 MQWs is EFISH. 40 pairs of amorphous Si–SiO_2 layers were evaporated by RF-sputtering on the vicinal (100) silicon wafer (Fig. 1). The parameters of radiation and sample are shown in Fig. 2. The dependence of the SHG intensity on the azimuthal angle and on the dc-electric field applied to the MQWs has been studied in transmission through the multilayered structure and in parallel polarizations of the fundamental and SHG wave. Azimuthal dependence reveals a two-fold symmetry and is completely anisotropic. The dependence of the SHG intensity on the dc-electric field applied to the MQWs presented in Fig. 2 has been studied in maximum of anisotropic dependence.

This oscillatoric dependence is interpreted as the feature of the screening of dc-electric field in layered structure of the MQWs. Note that classical screening gives essentially monotonic dependence, which is inconsistent with the experimental data. The electrostatic problem is solved in assumption that there is no current through the sample. Nevertheless, the electrons are supposed to come in wells due to the tunneling. Discrete analogue of the Poisson equation in two-dimensional case with boundary conditions is used to obtain the the electrostatic potential $\varphi(n)$ as function of a discrete variable n, which numbers n-th quantum well and the dependence of the electrostatical field E_0 on the voltage applied to the MQWs U. It is shown that the dependence of the electrostatical field E_0 on the voltage U have got oscillatoric behavior when $d + D\frac{\varepsilon_1}{\varepsilon} > \varepsilon a_0$, where d and D — thickness of the Si and SiO_2 layers, respectively, ε and ε_1 are their static dielectric constants, a_0—Bohr

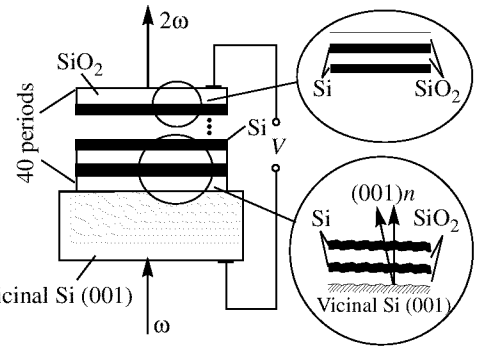

Fig. 1. Structure of the sample and geometry of experiment.

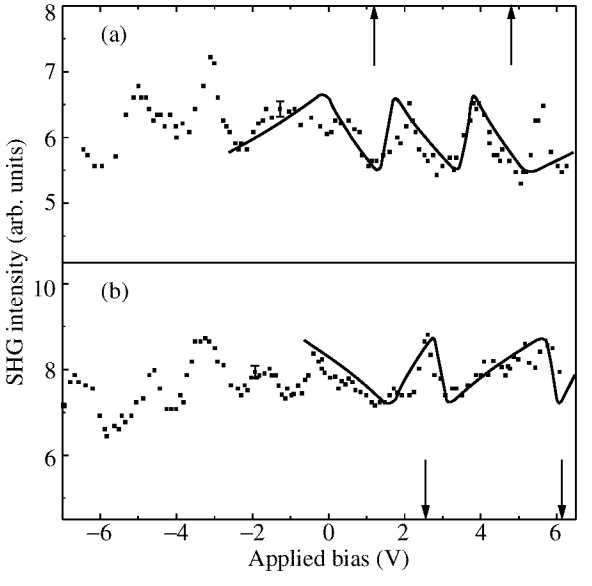

Fig. 2. The SHG intensity as a function of voltage. Experiment—square. Theory—solid line. Parameters of samples and radiation: (a) $d_{\mathrm{Si}} = 1.1$ nm, $D_{\mathrm{SiO_2}} = 3$ nm, $d_{\mathrm{Si}(001)} = 10^5$ nm, (b) $d_{\mathrm{Si}} = 1.1$ nm, $D_{\mathrm{SiO_2}} = 5$ nm, $d_{\mathrm{Si}(001)} = 10^5$ nm; $\lambda_\omega = 1064$ nm. Arrows indicate the region of validity of the model.

radius. This oscillatoric dependence $\varphi(n)$ appears due to excluding of the self-action in the Tomas–Fermi description. The screening in the single well is described using Hartree approach.

The azimuthal anisotropic dependence allows to assume that internal layers of the MQWs possess the small initial miscut (Fig. 1) with $2/m$ symmetry. However the external layers possess ∞/m symmetry. For the transmission geometry only $2/m$ layers contribute to the EFISH response. In this assumption the theoretical dependence of the SHG intensity on the external bias is obtained. Fig. 2 shows this dependence. The arrows indicate the region of validity of the model. The fit reveals good agreement to experimental data.

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Polarization anisotropy of photoluminescence of oxidized silicon nanocrystals

O. M. Sreseli†, D. I. Kovalev† and G. Polisski‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Department für Physik E16, Universität München,
D-85747 Garching, Germany

Abstract. Photoluminescence polarization and its anisotropy from oxidized silicon nanocrystals has been investigated.Unusual polarization anizotropy was observed under excitation with linear polarized light and large differences between excitation and detection energies. Oxide shell is shown to produce some effects enhancing the probability of no-phonon quasi-direct radiative transitions in nanocrystals. The observed anisotropy is explained by the anisotropy of heavy hole exciton in a quantum box.

Introduction

Porous Si is known to have a good memory of the linear polarization of the light used to excite the photoluminescence (PL).A review of this effect can be found in [1]. It has been established that polarization is due to an axial nanocrystallite (NC) asymmetry. For the colloidal particles having a spherical shape the polarization is totally absent [2]. In elongated nanocrystals (NCs), the anisotropy may have two origins, the electron confinement [3] or the effect of the depolarizing field created by the light-induced charges on the interfaces [4, 5]. Because of the difference in the optical dielectric constants inside and outsite the NCs, a depolarizing field appears when NCs are excited by light. It reduces the electric field inside the NCs, and the reduction is the smallest along the long NC axis. Light absorption is the largest in the NCs with the long axes aligned in the direction of the electric field of the exciting light. The excited NCs emit light also with preferential polarization along the long axis. The PL from the ensemble of randomly oriented NCs becomes thus polarized along the direction of linear polarization of the exciting light. Polarization degree of PL is isotropic in a sample plane, and anisotropy of polarization may appear only in the presence of anisotropic distribution of elongated NCs. For example, the artificially induced anisotropy of the NC distribution is reported in [6]. The anisotropy due to electronic confinement in elongated NCs seems to be effective for quasiresonant excitation of PL. The polarization studies achieved in these conditions reveal the symmetry of the ground states in Si NCs [7].

In this paper we study the PL polarization of oxidized silicon NCs prepared by thermal oxidation of porous-Si layer. We present an unusual behavior of the PL polarization at both non-resonant and quasiresonant PL excitation by linear polarized light. Appearence of polarization anisotropy in these cases is discussed in terms of dielectric model and quantum confinement.

1 Experiment

The porous-Si layers were made from the (100)Si wafers by conventional technique, the details of the sample preparation can be found elsewhere [8]. Oxidation was carried out in

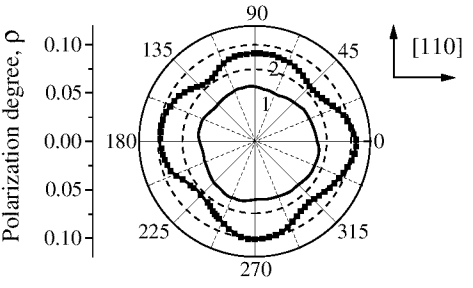

Fig. 1. Polar diagram of the PL polarization in porous Si prepared on (100)substrate. $\lambda_{ex} = 442$ nm (2.8 eV), $\lambda_{det} = 650$ nm (1.9 eV). (1) before oxidation; (2) after oxidation at 200°C, 5 min.

oxygen atmosphere at 200°C. The PL was excited by the linearly polarized light from the He-Cd laser (442 nm—non-resonant excitation) or from He-Ne laser (633 nm—resonant excitation). The polarization of the exciting light, $\vec{e_{ex}}$ lies in the surface plane of the layer, and its orientation with respect to crystalline axis can be changed by rotating the sample. The polarization degree is defined as

$$\rho = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$$

where $I_{\parallel}(I_{\perp})$ is the PL intensity polarized parallel (perpendicular) to $\vec{e_{ex}}$.

Measurements were done at 300 K. Details of setup are ascribed in [7].

2 Results and discussion

Angular dependences of the PL polarization under non-resonant excitation of the sample before and after oxidation are shown in Fig. 1. The isotropy of ρ in the (100) plane of porous layer (curve 1) was shown in [1, 5] and indicates the random orientation of the luminecing NCs in this plane.

After oxidation one may expect two effects: the reduction of NC sizes and, in the case of homogenious oxidation, the increase of their asymmetry (the ratio of long to short axis dimensions becomes larger). We see the first effect as shifting of PL spectrum to short-wave region and the second effect causes the enhancement of ρ (compare curve 1 and 2 in Fig. 1). When (100) plane is excited after oxidation procedure, the polarization anisotropy appears (curve 2 in Fig. 1). This is an unusual phenomenon for non-resonant excitation, since there is a great energy difference between exciting and emitting photons. We have studied this anisotropy at different wavelengths of detection (Fig. 2(a)). The polarization memory is higher when $\vec{e_{ex}}$ is parallel to [110] direction and has a minimum when $\vec{e_{ex}}$ is parallel to [100]. With increasing of the wavelength the degree of polarization decreases, but its anisotropy becomes more pronounced. The polarization anisotropy may be induced by structural anisotropy of NCs in case of different oxidation rates along different crystallographic axes. But the rates depend strongly on oxidation conditions. It is not straightforward to extent the data obtained for flat and polished crystallographic planes of c-Si on arbitrary NC curved surfaces.

Before discussing the second possible reason—electronic confinement—we investigated polar diagrams of PL polarization at resonant PL excitation (Fig. 2(b)). Resonant excitation of oxidized samples gives enhanced PL polarization degree and pronounced polarization anisotropy which does not decrease with increasing λ_{det}. Again ρ is higher in

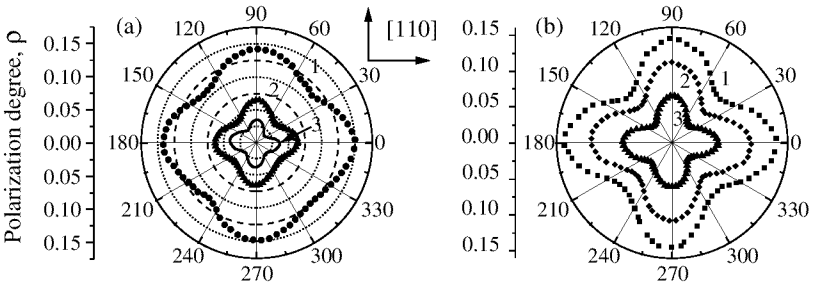

Fig. 2. Polar diagrams of the PL polarization in oxidized Si NCs under non-resonant (a) and resonant (b) conditions; (a) $\lambda_{ex} = 442$ nm (2.8 eV), λ_{det}: 1—550 nm (2.25 eV); 2—700 nm (1.77 eV); 3—750 nm (1.65 eV); (b) $\lambda_{ex} = 633$ nm (2.8 eV), λ_{det}: 1—670 nm (1.85 eV); 2—700 nm (1.77 eV); 3—750 nm (1.65 eV).

[110] direction. Under resonant conditions the dependence of ρ on the crystalline axes reflects the warping of the heavy hole sub-band [7]. Under resonant excitation, the absorption takes place in states which are close to the ground state of the exciton. In a quantum box the ground state would have a nature of the heavy holes because their confinement is smaller. Along the [110] direction the heavy hole mass is larger than along the [100] direction. For the same NC size, the absorption will be higher in the [110] aligned NC, because in the quantum box the density of states is higher along this direction. This anisotropy must dissapear for detection energies far from the excitation ones. In [7] this occurs at differences $\Delta = E_{ex} - E_{det}$, equal to 0.2–0.3 eV. At higher Δ, for lower detection energies, contributions from light hole excitons become important. In the light hole band the mass along [100] is larger than the mass along [110], and ρ becomes greater in [100] direction [7, 10]. In our experiments anisotropy of polarization memory is well-defined till $\Delta = 0.5$ eV (setup limitation), not to mention anisotropy under non-resonant excitation at Δ, equals to 1–1.5 eV. We explain our results by enhancement of no-phonon quasi-direct radiative transitions. It was shown theoretically [11] and experimentally [12] that in smaller Si NCs the probability of no-phonon transition increases with respect to phonon-assisted processes. For confinement energies above 0.7 eV the no-phonon transitions begin to dominate. Oxidation, first, increases the confinement energy due to reduction of NC sizes, second, increases asymmetry of NC and localization of the exciton along its longer axis. For the same confinement energies no-phonon transitions are some times stronger in Si NCs having a SiO_2 cover [12]. So even at large Δ we see anisotropy of polarization due to asymmetry of Si heavy hole band. We assume also that larger NCs oxidize with greater rates than the smaller ones and have larger SiO_2 cover. This explains why at one excitation energy ρ does not decrease with increasing λ_{det} (increasing Δ).

Thus, we have observed polarization anisotropy in photoluminescence of Si oxidized NCs and explained it in frame of quantum confinement model.

Acknowledgements

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